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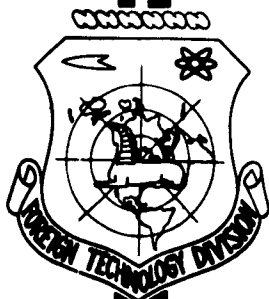
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(SELECTED ARTICLES)

FOREIGN TECHNOLOGY DIVISION



AIR FORCE SYSTEMS COMMAND

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Calculating Thermodynamic Properties of Real Gases at High Temperatures

by

A. G. Tabachnikov

Described is a method of extrapolating by temperatures of virial coefficients. The method was used for formulating an equation of the state of nitrogen in the range of temperatures of from 0-3000°C.

Contemporary development of a number of branches of science and technology needs data on the thermodynamic behavior of substances, particularly gases, in the zone of high temperatures.

The presently available experimental data on thermodynamic properties of gases are in a majority of instances limited to temperatures of 150-200°C. In recent years efforts are being made to experiment at much higher temperatures [1, 2]. But unfortunately, even these single experiments do not go beyond temperatures of over 1000°C.

Perfectly natural are therefore efforts to determine thermodynamic properties at high temperatures, on the basis of experimental in the zone of moderate temperatures [3, 4, 5].

Such calculations are mostly made with the aid of the virial equation of state

$$PV = RT \left[1 + \frac{B(T)}{V} + \frac{C(T)}{V^2} + \dots \right] \quad (1)$$

for the calculation of coefficients of which there are definite theoretical hypotheses.

The fact is, statistical physics offers the principal possibility of calculating all so-called virial coefficients of equation (1) if the energy of paired reaction of molecules $\epsilon(r)$ is known. At present time for a number of proposed interaction potentials $\epsilon(r)$ were made preliminary calculations, necessary to determine the second $B(T)$ and third $C(T)$ of virial coefficients [6].

But it should be pointed out that of all virial coefficients sufficiently reliab-

le can be calculated only the second ($B(T)$) which is relatively less sensitive to the form of interaction function.

In the case of much higher virial coefficients the proposed interaction potentials give less satisfactory results.

In connection with this it was decided to determine the virial coefficients of equation (1), on the basis of theoretically calculated second virial coefficient $B(T)$ and rules, emanating from the equation of state, introduced by Ya. Z. Kazavchinskiy [7].

$$PV = \alpha_0 + \alpha_1 T + \beta \psi + \gamma \psi^2 + \dots, \quad (2)$$

where $\alpha_0 = \sum_{i=1}^n \frac{a_i}{V^i}$; $\alpha_1 = R + \sum_{i=1}^n \frac{b_i}{V^i}$; $\beta = \sum_{i=1}^n \frac{c_i}{V^i}$; $\gamma = \sum_{i=2}^n \frac{d_i}{V^i}$; ψ - mono-

ψ - monotonously decreasing function of temperature, the form of which will be discussed below; n depends upon the described range of densities.

The selection of equation (2) to solve the given problem does not appear to be accidental. The fact is, that fully agreeing with the Bogolyubov-Mayer [8] general equation of state, it will by its form very accurately reflect all behavioral characteristics of real gases. The structure of equation (2) is such, that it is possible to use same for describing practically any one parameter change zone. It should be pointed out, that in form (2) were formulated equations of state for a greater number of gases varying in their qualities [9 - 18], the parameters of which varied in densities from zero to two and more critical, and in temperatures - from saturation curve to temperatures, considerably exceeding the Boyle temperature.

The given considerations allow to assume, that not only equation (2) in itself, but also the relations obtained from it appear to be valid in the entire zone of parameter change, including also at high temperatures.

Having transformed equation (2) into form (1) we will obtain for virial coefficients the following expressions:

$$\begin{aligned} B(T) &= b_2 + \frac{a_2}{RT} + \frac{c_1 \psi}{RT}, \\ C(T) &= b_3 + \frac{a_3}{RT} + \frac{c_2 \psi}{RT} + \frac{d_2 \psi^2}{RT} + \end{aligned} \quad (3)$$

As is evident from the mentioned expressions, if in the investigated range of ^{temperatures} ψ is known the function ψ , then the determination of all virial coefficients is reduced to finding by experimental data the constants $a_1, b_1, c_1, d_1, \dots$ in expressions (3), (4).
 ...The equation of state compiled in such a ————— way will be valid, apparently, in this range of temperatures, in which the temperature function ψ was determined. In turn the temperature function ψ in a sufficiently broad range of temperatures can be principally separated from the theoretically calculated second virial coefficient.

This defines the method of formulating the equation of state suitable for calculating at high temperatures, which consists in the following:

1. On all experimental isothermal curves are separated virial coefficients, the number of which depends upon the described range of densities. Separation of virial coefficients can be brought about by the known method of plotting and extrapolating in the zero density of isotherms in coordinates

$$V \left(\frac{PV}{RT} - 1 \right) - \frac{1}{V} \cdot (4a)$$

$$V^2 \left(\frac{PV}{RT} - 1 - \frac{B}{V} \right) - \frac{1}{V} \cdot (4b)$$

to determine the corresponding second $B(T)$, third $C(T)$ and so on, by the virial coefficient.

2. By the experimental values of the second virial coefficient $B(T)$ are determined the parameters of the accepted interaction ————— potential and to calculate the values $B(T)$ in the investigated range of temperatures. Selection of the interaction potential is determined in each case by — the properties of the gases.

3. From the theoretically calculated second virial coefficient is separated the temperature function.

4. Having to our disposal the temperature function ψ and table separated, on all experimental isotherms, virial components, by the method of least squares we find the constants $a_1, b_1, c_1, d_1, \dots$ in equations (3), (4)....

It still remains to be explained:

1. Method of separating temperature function ψ from second virial coefficient.

2. Necessary number of analysis members in so-called curvilinear part of equation

(2) ($\beta\psi + \gamma\psi + \dots$).

In response to the first question we will turn to the theoretical equation of the second virial coefficient [19]

$$B(T) = 2\pi \int_0^\infty \left\{ 1 - \exp \left[- \frac{\epsilon(r)}{kT} \right] \right\} r^2 dr \quad (5)$$

It is necessary to assume, that there is a certain minimum permissible distance between molecule r_0 , because at $r < r_0$ $\epsilon(r) = \infty$. It can then be written

$$B(T) = \frac{2}{3} \pi N r_0^3 + 2\pi N \int_{r_0}^\infty \left\{ 1 - \exp \left[- \frac{\epsilon(r)}{kT} \right] \right\} r^2 dr \quad (6)$$

Having broken down $\exp \left[- \frac{\epsilon(r)}{kT} \right]$ into series according to $\frac{1}{T}$ and integrating termwise, we will obtain

$$B(T) = k_0 + \frac{k_1}{T} + \frac{k_2}{T^2} + \dots \quad (7)$$

It is evident from the obtained expression that to determine the temperature function ψ is necessary the second virial coefficient, theoretically calculated to sufficiently high temperatures (desirable to temperatures, corresponding to its maximum, or over), analytically represented in form (7), then, as is evident from comparing (7) and (3) the temperature function ψ with an accuracy to constant multiplier $\frac{C_1}{R}$ will be equal to

$$\psi = \frac{k_1}{T} + \frac{k_2}{T^2} + \dots \quad (8)$$

To offer an answer to the second question we will state, that by its physical nature the curvilinear part of the equation (2) takes into consideration the interaction of associated molecules [8] and, consequently, the basic role is played in the zone of low temperatures, particularly near the saturation curve, consequently the number of necessary decomposition members depends here upon the temperature range of experimental data, on the basis of which the constants of equation (2) are determined.

On the other hand, in connection with the fact, that by form equation (2) at $V = \text{const}$

represents an isochore equation, it is possible to evaluate the nature of its curvilinear part by the isochore configuration features of real gas.

Analysis of real gas isochores shows :

- 1) Up to critical density ($d < d_k$) isochores have a curvature of one sign over the entire range of temperature changes;
- 2) after critical density the sign of isochore curvature changes, but not over the entire range of temperatures, but only in the range of from T_5 to $\gamma = T/T_k \approx 1.5 - 1.7$. At temperatures greater than $1.7 T_k$, the curvature sign remains as before. In other words, at $d > d_k$ the isochores appear to be lines of double curvatures;
- 3) when $d \geq 2d_k$ the isochores again have a curvature of one sign, the very same sign as at $d < d_k$.

It is evident from this analysis, that in order to describe with equation (2) the temperature range, beginning with a saturation temperature, it is necessary to have a minimum of two members in the curvilinear part ($\beta\psi$ and $\gamma\psi^2$), whereby, apparently, at $d > d_k$ β and γ should have different signs.

If constant equations of state are determined by experimental data at temperatures, higher than $1.7 T_k$, where all isochores have a curvature of one sign, it is possible to confine oneself only to the first stage of temperature function ψ .

In the proposed method in the process of separating virial coefficients — and binding same with second virial coefficient $B(T)$ is attained simultaneously a graphic and simple agreement of experimental data.

We will discuss below the compilation of an equation of state for nitrogen.

Equation of State of Nitrogen

In the range of temperatures from 0 - 800°C ^{for} the second virial coefficient of nitrogen data are already available [2,20-22]. At temperatures below 0°C the values of the second virial coefficient were determined by us by PVT-data by Holborn, Otto and Kamerling, Onnes and Yurk [23]. As result of graphic smoothing were obtained

mean values of the second virial coefficient in the temperature range of 125-800°C (table 1).

To select the form of interaction function of molecules $\varphi(r)$ the experimental values of the second virial coefficient were compared with theoretically calculated through potentials (6-12) Lenard-Johns and (exp.6) Buckingham and by the Kilhar equation (table 2)

For the case of the Lennard-Johns (6-12) potential

$$\varepsilon(r) = 4\varepsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right] \quad (9)$$

calculation of the second virial coefficient is done by formula

$$B(T) = -\rho F(\tau) \quad (10)$$

to which after integration is brought equation (5), if in role of interaction function is accepted expression (9), whereby

$$\rho = \frac{2}{3} \pi N \sigma^3, \quad \text{and} \quad \tau = \frac{kT}{\varepsilon} \quad (11)$$

Function $F(\tau)$ for $0.3 \leq \tau \leq 400$ tabulated in experiment [24].

Table 1. Experimental values of second virial coefficient of nitrogen
($B \cdot 10^3$, Amagat units, $V_0 = 22404 \text{ cm}^3/\text{mol}$)

1) t°C	2) Holborn & Otto	3) Michellie & Assoc.	4) Sorel	5) OIIMF	6) Average
1)	2)	3)	4)	5)	6)
-125				-8.460	-8.460
-100				-2.260	-2.260
-75				-1.610	-1.610
-50				-1.157	-1.125
-25				-0.743	-0.755
0	-0.467	-0.460			-0.460
25		-0.209			-0.213
50	-0.023	-0.013			-0.020
75					0.145
100	0.269	0.293			0.290
150	0.514	0.548	0.5038		0.509
200	0.686		0.6721		0.672
300	0.909		0.9096		0.910
400	1.047		1.0643		1.064
500			1.1705		1.171
600			1.2486		1.249
700			1.3061		1.306
800			1.3478		1.348

Table 2. Comparison of Experimental Values of Second Virial Coefficient of Nitrogen with Calculated
($B \cdot 10^3$, Amagat units, $V_0 = 22404 \text{ cm}^3/\text{mol}$)

$t, ^\circ\text{C}$ 2) Experimental 3) Lennard-Johns 4) Buckingham, $a=13$, 5) Kikhar, $\epsilon/k = 124^\circ\text{K}$.
 $\epsilon/k = 94.75^\circ\text{K}$, $\epsilon_m/k = 91.21^\circ\text{K}$ $r_0 = 3.43 \text{ \AA}$
 $\sigma = 2.846$ $b_0 = 4.406$

1	2	3	4	5
-125	-3.266	-3.131	-3.221	-3.360
-100	-2.260	-2.222	-2.291	-2.370
-75	-1.610	-1.580	-1.628	-1.687
-50	-1.125	-1.101	-1.135	-1.184
-25	-0.755	-0.726	-0.756	-0.791
0	-0.460	-0.436	-0.457	-0.484
25	-0.113	-0.188	-0.216	-0.228
50	-0.020	-0.022	-0.015	-0.022
75	0.145	0.192	0.161	0.141
100	0.290	0.330	0.292	0.280
150	0.509	0.533	0.519	0.500
200	0.672	0.693	0.684	0.681
300	0.910	0.928	0.921	0.925
400	1.064	1.085	1.072	1.084
500	1.171	1.192	1.176	1.194
600	1.249	1.267	1.249	1.275
700	1.306	1.312	1.300	1.334
800	1.348	1.364	1.339	1.383

The values σ and ϵ/k (table 2) were determined by experimental values of the second virial coefficient by the Lennard-Johns method [25].

Buckingham [26] introduced an interaction potential, in which theoretically more founded in comparison with (9) is the member, considering the repulsion energy. One of the modifications of this potential appears to be the so-called (exp-6) potential.

$$u(r) = \frac{\epsilon_m}{1 - 6/\alpha} \{ (6/\alpha) \exp[\alpha(1 - r/r_m)] - (r_m/r)^6 \}, \quad r > r_{\max}, \quad (12)$$

$$u(r) = \infty \quad \text{при} \quad r < r_{\max}. \quad (13)$$

The idea of constants ϵ_m , r_m and r_{\max} is plain from fig.1; α determines the curvature of the repulsion potential and is connected with the value of in termolecular distance r_0 , at which $u(r) = 0$.

Equation (5) can be reduced to form

$$B(T) = b_0 \Phi(\alpha, \tau), \quad (14)$$

where

$$b_0 = \frac{2}{3} \pi N r_m^3, \quad \tau = \frac{kT}{\epsilon_m} \quad 14a$$

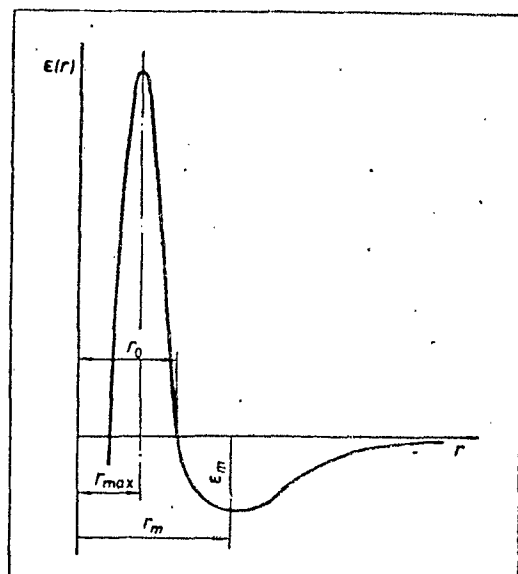


Fig.1. Graph, illustrating idea of constants ξ_m , r_m , r_{max}

The function $\phi(\alpha, \tau)$ for seven values α in limits $0.4 \leq \tau \leq 400$ is tabulated in report [27].

Parameters α , b_0 and ξ_m/k were determined by the above described Lennard-Johns method with the only exception that the presence of the third parameter α leads to the necessity of plotting a series of curves $\lg|\phi(\alpha, \tau)| - \lg \tau$ for various α values.

By analyzing table 2 it can be concluded, that, first of all, in case of nitrogen the consideration of the nonspherical form of its molecule does not produce a considerable effect, on the other hand, all calculated values of the second virial coefficient are close to each other, a certain discrepancy is being observed only at high temperatures.

In connection with the fact, that with a rise in temperature there is an intensified influence of the repulsion potential, for the calculation of the second virial coefficient at high temperatures was accepted the Buckingham potential.

On the basis of formula (7), by the method of the least squares was compiled equa-

tion of the second virial coefficient, valid in the temperature range of from 125-3000°C

$$10^3 B = 1,2303 + \frac{8,152}{\theta} - \frac{104,5}{\theta^2} + \frac{388,7}{\theta^3} - \frac{808,8}{\theta^4} + \frac{865,1}{\theta^5} - \frac{373,5}{\theta^6} \quad (14c)$$

where B-in Amaga units; $\theta = \frac{T}{100} ^\circ K$.

When compiling equation of state we used experimental data [29 and 2] in the range of temperatures of from 0 - 800°C.

For convenience the equation of state was compiled not in virial form (1) but in a somewhat changed form

$$PV = RT + B_p \rho + C_p \rho^2 + \dots \quad (15)$$

Plotting of isotherms $A = \frac{1}{\rho^2} (PV - RT - B_p \rho)$ in dependence upon ρ^2 showed, that up to $\rho = 360$ Amaga units within limits of experimental error the experimental points are plotted on straight lines. This indicates, that in the range of densities 0 - 360 $\frac{\text{Amaga} \cdot \text{g}}{\text{units}}$ equation (15) can be presented in form of

$$PV = RT + B_p \rho + C_p \rho^2 + D_p \rho^3 \quad (15a)$$

whereby it becomes apparent that

$$C_p = \lim_{\rho \rightarrow 0} A, \quad D_p = \left(\frac{\partial A}{\partial \rho^2} \right)_{T, \rho \rightarrow 0} \quad (16)$$

The C_p and D_p values determined in this way were reflected by an analytical method of least squares in form of

$$C_p = a_1 + b_1 \theta + c_1 \psi, \quad D_p = a_2 + b_2 \theta + c_2 \psi \quad (16a)$$

where ψ is determined from equation of the second virial coefficient in conformity with (7) and (8).

Finally for densities of 0 - 360 Amaga units and temperatures of from 0-3000°C the equation of state has the form of

$$PV = R \theta + B_p \rho + C_p \rho^2 + D_p \rho^3 \quad (16b)$$

where

$$10^3 B_p = 2,986 + 0,4506 \theta + \psi \quad (17)$$

$$10^6 C_p = 10,164 + 0,1955 \theta - 1,6701 \psi \quad (18)$$

$$10^9 D_p = 0,6177 + 0,0740 \theta + 0,1114 \psi \quad (19)$$

$$\psi = \frac{88,27}{\theta} - \frac{142,4}{\theta^2} + \frac{296,2}{\theta^3} - \frac{316,8}{\theta^4} + \frac{136,8}{\theta^5} \quad (20)$$

In the equations were adopted: P - in phys. atmospheres; V and ρ - in Ångström units;
 $V_0 = 22404 \text{ cm}^3/\text{mol}$; $R = 0.36625 \text{ degrees}^{-1}$.

A check of equation in accordance with experimental data [29,2] showed that the mean error is 0.05%, and the maximum - does not exceed 0.16% (the check was made on 103 points).

Abbreviations

ϵ_r - energy of interaction of two molecules, situated at a distance r from each other; k - Boltzmann constant; N - Avogadro number; ϵ - value of potential curve minimum;
 σ - intermolecular distance, at which $\epsilon(r) = 0$.

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Leakage of Gas into a Liquid through the Laval Nozzle

by

M. G. Moiseyev

Leakage of a gas stream into a liquid was investigated before by [1] [2]. But in these experiments was investigated the discharge of a subsonic air stream into water. The results, given in this report, appeared to be the development and expansion of these investigations over the zone of much higher discharge rates. The experiments were made on a special installation. The discharge of the stream was realized horizontally into a tank, filled with water. The level of the water column above the cut of the nozzle was 300 mm. To assure photographing of the stream the tank was provided with windows, covered with transparent organic glass.

It should be mentioned, that the floating stream passes basically only there, where the pressure is already low. This is confirmed by measurements and photos. In this way, the effect of gravity on the outflow of a supersonic gas stream in horizontal plane within limits of the investigated section is practically null.

To measure pressure in the stream was used as specially prepared Pitot tube with electric heating. The tube moved in horizontal plane with the aid of a coordinator. Heating of receiving tube, was found necessary, because without it the water froze, and measurements were impossible.

In the process of experimental investigation was measured total pressure along the axis and in transverse direction. The investigation was made at various flow velocities at the output from the nozzle and various unforeseen things. Under unforeseen things we will understand the pressure ratio in the output section of the nozzle

to the pressure in the surrounding medium. Four nozzles with different M -number at output were used: $M_{out} = 1.73; 2.09; 2.58; 3.00$. The diameter of the critical section of all nozzles was 4 mm, angle of opening - 10° .

Measurements along the axis of the stream were made at a distance of up to 20-35 calibers of output section of nozzle. Near the nozzle, within limits of 3-4 calibers, measurements were made within 1 mm, then within

2 mm, and further on the basic section of the stream within larger intervals.

Across stream measurements we realized within 1 mm.

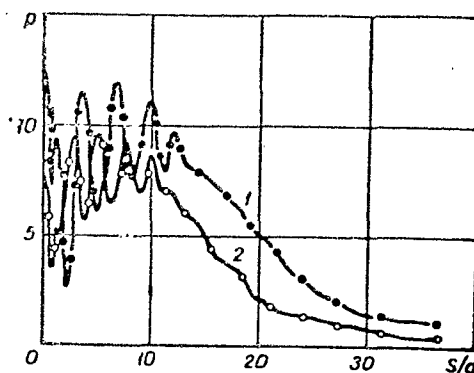


Fig.1. Change in pressure p (gage atm) measured by Pitot tube, along the axis of the stream ($M_{out} = 2.58$); 1- $n=1.5$; 2- $n=1.0$

in graphs. By studying the curves plot

ted in fig.1 it is evident, that in a gas stream, being discharged into water, should be distinguished as during the discharge into the air, two sharply differing sections. This first of all, is the initial section of the stream, in which sharp pressure fluctuations are observed, measured by the Pitot tube. The length of this section is sufficiently large and it grows with the increase in incompressibility (unforeseen things). On the other hand, the basic section, on which a smooth pressure drop is observed, measured by the Pitot tube.

A comparison of measurement results shows (fig.2) that also the nature of pressure fluctuations along the axis of the stream, and their value, and length of the entire initial section of air supersonic stream, flowing into water and into air, are in quite accurate conformity. This circumstance indicates, that within the limits of the initial section of the air flow propagating in water, is retained the very same configuration of shock waves, as in a stream, being discharged into the air. And this

indicates, that within the limits of the initial section there is no intensive mixing of two phases, otherwise the affinity (similarity) with the stream, spreading in air medium, would be disrupted.

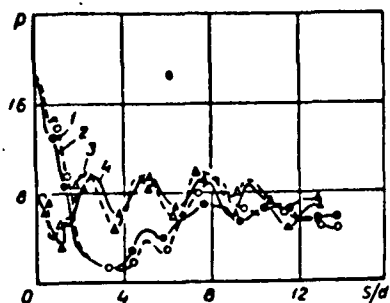


Fig.2. Comparing results of measuring pressure p (gauge atm) along axis in initial section of stream in water and in air (diameter of critical section of nozzle 8 mm);

1- $M_{out} = 1.96$; $n = 3.22$ (in air); 2-2.09; 3-2.49; 1.0 (in air); 4-2.58; 1.0 (in water)

Measuring pressure along the profile of the stream shows, that along the axis of the stream in the zone of high pressures no sharp expansion is observed. This circumstance also confirms the analogy of initial sections of supersonic air streams, spreading in the air and in water. A sharp expansion of the

stream, observed on photos, and also the wide zone of lower total pressures along

the periphery of the stream indicate, that the air stream, spreading in water, has a broad

boundary layer, consisting of gas-liquid mixture, which moves at slow pace in comparison with the velocity of the central stream. The angle of stream expansion is $18-25^\circ$ where- by a greater angle value corresponds to greater values of incalculability.

On the basis of above findings it can be recommended to calculate the initial section, with the exception of the boundary layer, for the supersonic air stream, flowing into water, as well as for the stream, flowing into an air medium.

On the basic section of the stream the pressure measured along the axis decreases smoothly. The pressure fields, obtained by the Pitot tube, were processed in dimension less coordinates

$$\frac{\Delta p}{\Delta p_m} = f\left(\frac{y}{y_0}\right). \quad (//)$$

In fig.3 is given the field of pressures, measured by the Pitot tube, on the basic section of air stream in water. Here are plotted experimental points, corresponding to various nozzles, conditions and distances from the cut of the nozzle.

Results of experiments are in quite excellent conformity with the solution, obtained by G.N. Abramovich [1] for the problem of the gas stream in a liquid according to which the profile of the kinetic head is approximately described by dependence

$$\frac{\Delta p}{\Delta p_m} = (1 - \xi^{1.5})^2, \quad (2)$$

where $\xi = y/b$.

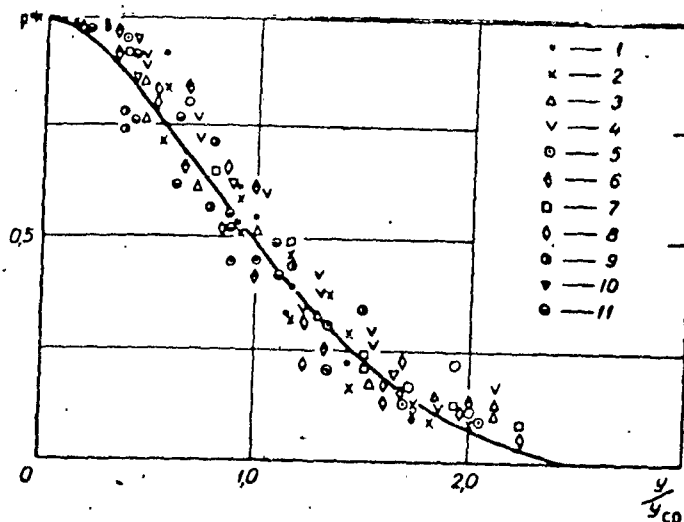


Fig. 3. Field of measured pressures on basic section of stream

$$p^* = \frac{\Delta p}{\Delta p_m} = f\left(\frac{y}{y_{cp}}\right):$$

1-M_{out}=3; n = 1; S/d=11.2; 2-3; 1; 13; 1; 3-3; 1; 16.6; 4-3; 1; 19.5; 5- 3; 0.5; 7.8; 6- 3; 0.5; 10.9; 7- 2.58; 0.5; 10.0; 8- 2.58; 1; 11.8; 9- 2.58; 1; 17.0; 10-1.73; 2; 9.2; 11- 1.73; 5; 18.8

Such a result for the basic section of subsonic air stream in water was obtained by B.F. Glikman [2].

It is necessary to mention the pulsating nature of the outflow. Pulsations are accompanied by shocks. During pulsations a part of the liquid in the nozzle is repulsed in direction, opposite of the direction of flow. Pulsation frequency is 0.5 - 1 c.

Designations

p - pressure, measured by Pitot tube; S - distance from nozzle out; d - diameter of output section of nozzle; Δp , Δp_m - excess pressure in given point and on axis of stream; y , y_{cp} - distance from given point and point, where $\Delta p = 0.5 \Delta p_m$, to axis of stream;

b-radius of stream.

Literature

1. Abramovich G.N; Theory of Turbulent Streams, Fizmatgiz, Moscow, 1960
2. Glikman B.F; Izvestiya Akademii Nauk SSSR; Otdel. Tekhn. Nauk; Energetika i Avtomatika, No.2, 1959

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